Pressure enhanced tunnel magnetoresistance in Co-Al-O granular films

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Effects of high pressure on tunnel transport and magnetoresistance have been investigated in $Co_{52}Al_{20}O_{28}$ insulating granular films. It is found that the temperature dependence of electrical resistivity $\rho(T)$ is affected strongly by applying pressure while the $T^{-1/2}$ dependence in $\rho(T)$ is still observed at high pressures. Furthermore, tunnel magnetoresistance is enhanced by more than 2% at 3.1 GPa. The results are discussed briefly on the basis of higher-order tunneling theory.

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I. INTRODUCTION

The giant magnetoresistance (GMR) was discovered in Fe/Cr magnetic multilayers by Baibich *et al.*[1] In various layered and granular systems, GMR and large tunnel magnetoresistance (TMR) have been observed to date.[2–12] The GMR and TMR effects have been investigated not only in the application but also in the fundamental aspects in the transport properties in materials. However, there have been no complete understanding both in GMR and TMR.

Since pressure is a good tool to change the electronic and structural properties continuously, a lot of new aspects can be expected in the research of transport properties at high pressure in these materials. In fact, the pressure effects on GMR of Fe/Cr or Co/Cu magnetic multilayers have been investigated extensively by the present authors (G.O. and K.T.), in which many important results were reported.[13–18] A Co-Al-O granular film is a typical example of granular materials exhibiting TMR,[9,10] which has a structure that Co granules (2–3 nm) are embedded in an insulating Al oxide. The conductance of granular systems shows a characteristic temperature dependence due to tunneling between Co granules across the insulating Al oxide. In this transport, the charging energy of Co granules and the distance between them have been well known to be crucially important. Therefore, significant pressure effect for TMR of Co-Al-O granular films is expected. In this paper, we report the effects of pressure on the electrical resistivity and magnetoresistance due to tunneling effect in granular systems.

II. EXPERIMENTAL PROCEDURE

Co-Al-O granular films were prepared on glass substrates using reactive-sputtering technique with a Co-Al alloy target and mixed gas of $Ar+O_2$.[19] The chemical composition of the film was determined by energy dispersive method in x-ray spectroscopy to be $Co_{52}Al_{20}O_{28}$ (hereafter we describe as 52 at. % Co), in which the volume fraction of Co was approximately 59 vol. %. The TEM observation and magnetization measurement revealed that the diameter of Co granules d was distributed to some extent. The mean diameter of granules $\langle d\rangle$ and the mean distance between granules $\langle s\rangle$ are estimated to be $\langle d\rangle=2-3$ nm and $\langle s\rangle=1$ nm or less.[20,21] Temperature dependence of electrical resistivity ρ and magnetoresistance (MR) were measured using a dc four-terminal method. The maximum applied magnetic field was 2 T. The current and magnetic field directions were always in plane. Hydrostatic pressure was generated up to 3.1 GPa by the piston-cylinder device utilizing the conventional teflon-cell technique. The pressure inside the cell was always kept constant by controlling the load of hydraulic press within $\pm 5\%$ throughout the measurement. The details of the present high-pressure apparatus were reported previously.[22] We also measured $\rho(T)$ curve of the specimen $Co_{43}Al_{24}O_{33}$ (43 at. % Co) for the comparison, which is away from the percolation limit compared with 52 at. % Co.

III. RESULTS AND DISCUSSION

A. Temperature dependence of the electrical resistivity

Figure 1 shows the temperature dependence of the electrical resistivity ρ under zero and high pressure up to 3.1 GPa. ρ increases with decreasing temperature. The overall behavior of $\rho(T)$ under high pressure is similar to that at 0 GPa (ambient pressure). Therefore the conduction mecha-

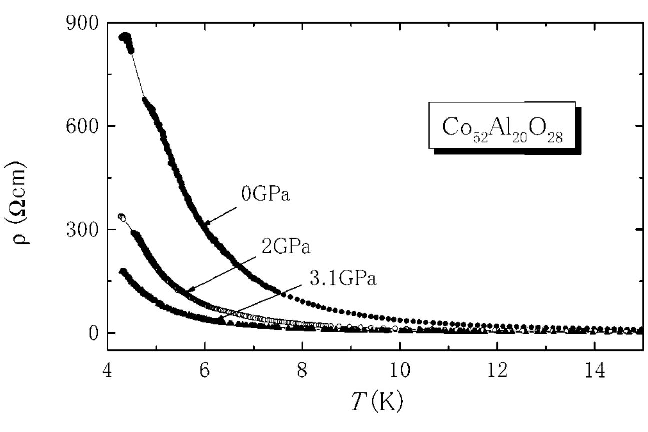

FIG. 1. The temperature dependence of resistivity under zero and high pressure for the $Co_{52}Al_{20}O_{28}$ thin film.

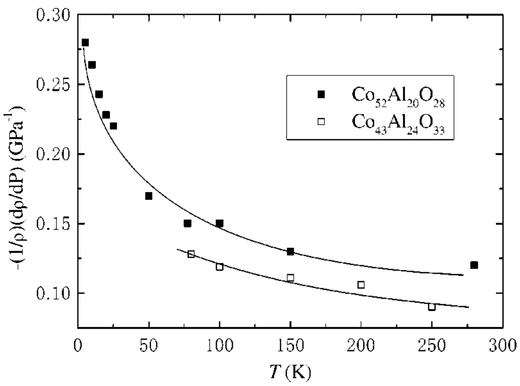

FIG. 2. The pressure coefficient of the electrical resistivity ρ as a function of temperature for $Co_{52}Al_{20}O_{32}$ (■) and $Co_{43}Al_{24}O_{33}$ (□), respectively.

nism under high pressure is almost the same as that at ambient pressure. But the increasing rate of resistivity $|d\rho/dT|$ becomes small as pressure increases. This indicates that pressure enhances the tunneling conduction at low temperature. The pressure coefficients $1/\rho(\partial\rho/\partial P)$ are easily estimated from the data shown in Fig. 1. The coefficients of 52 at. % Co is shown in Fig. 2 as a function of temperature. After releasing pressure, the magnitude of resistance was nearly same before pressurizing within the experimental error. It is seen that the coefficients increase slightly with decreasing temperature down to 100 K but shows a steep increase below 100 K. The pressure coefficients of 43 at. % Co are also shown in Fig. 2, which is found to be smaller than that of 52 at. % Co. The coefficients vs temperature curve of 52 at. % Co is nearly parallel to that of 43 at. % Co. The temperature dependence of the coefficients will be discussed later.

For the electrical resistivity of granular films, the following equation has been applied:[23,24]

$$\rho = A \exp\left\{2\sqrt{\frac{C}{k_B T}}\right\}, \tag{1}$$

$$C = \kappa s E_C,$$

where A is the constant, C is the activation energy, s is the tunnel-barrier thickness, E_C is the charging energy of metallic grain, and κ is the decay factor of tunnel probability, proportional to the square root of effective potential barrier height ϕ. From Eq. (1), we derive the equation $\ln\rho(T) \propto 2\sqrt{C/k_B}T^{-1/2}$. In order to examine this relationship, we plotted the experimental values of $\ln\rho$ as a function of $T^{-1/2}$, which is shown in Fig. 3. It is found that the present data can be approximately expressed by this relationship, although the values of the coefficient $2\sqrt{C/k_B}$ depend slightly on temperature range. The values of $2\sqrt{C/k_B}$ in the high-temperature range ($T \gtrsim 25$ K) are smaller than those in the low temperature ($T \lesssim 25$ K). It should also be noted that the linear relationship in $\ln\rho$ vs $T^{-1/2}$ is still observed under high pressure.

Figure 4 shows $2\sqrt{C/k_B}$ as a function of pressure. The values of $2\sqrt{C/k_B}$ were obtained by fitting the experimental data to Eq. (1) in the temperature range from 6 K to 25 K.

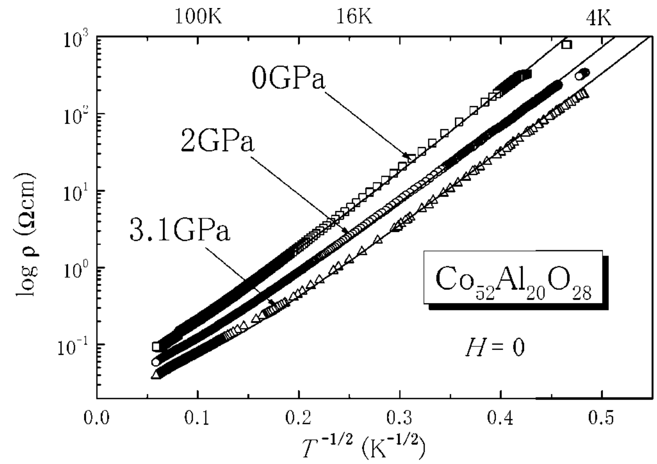

FIG. 3. $\ln\rho$ at 0, 2, 3.1 GPa as a function of $T^{-1/2}$. The solid lines are the fitting curves to Eq. (3) by assuming. $E_C/k_B = 23$ K, $\kappa\langle s\rangle = 4$, 3.26, and 2.95 for 0, 2, and 3.1 GPa, respectively. See the text for the details.

We measured for two different samples, A and B, having almost the same composition (52 at. % Co) in order to check reproducibility of the data, because a compositional fluctuation may exist in the sputtered film. The coefficients are found to decrease approximately linearly with increasing pressure: $2\sqrt{C/k_B}$ decreases by about 11% near 3 GPa. There is no large difference between two samples except the absolute value of C. In a simple consideration, the Co granules and Al-oxide matrix are compressed in the similar way under high pressure. $C(\sim sE_C)$ is expected to be constant, since E_C is roughly proportional to $1/d$, i.e., $C \sim s/d$, and both s and d decrease at the same rate, where d is the diameter of Co granules. However, the observed pressure dependence of C is quite different from this simple idea mentioned above. From $C = \kappa s E_C$, the experimental result suggests that the Al-oxide matrix is more compressible than Co granules and the remarkable change in C is mainly due to the decrease of s. With decreasing s, $\kappa(\sim\sqrt{\phi})$ may also be reduced. The pressure coefficient of $2\sqrt{C}$ is $\simeq -7\%$ for the high temperature range (25 K–room temperature), which is twice larger than that of $Co_{43}Al_{24}O_{33}$ (43 at. % Co), $\sim -3\%$, as was reported previously.[25] The 43 at. % Co is away from the percolation

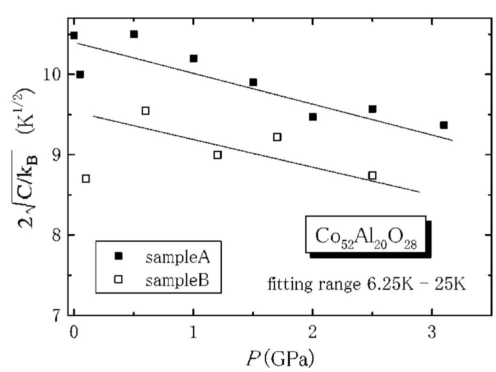

FIG. 4. The coefficient of $2\sqrt{C/k_B}$ as a function of pressure for $Co_{52}Al_{20}O_{28}$ samples, A and B. The solid lines are guide to the eye for each sample.

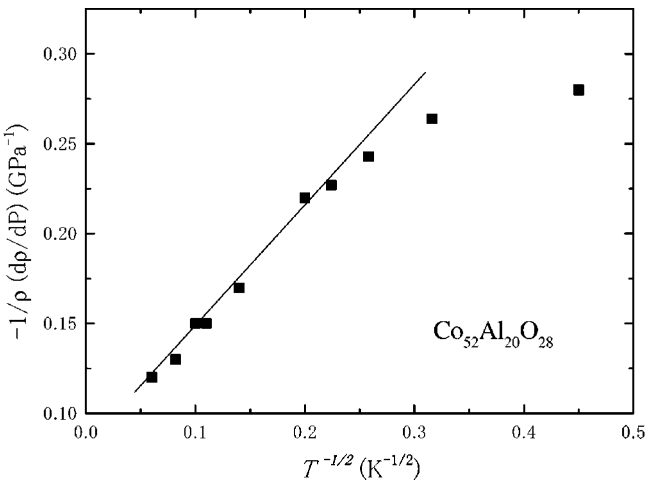

FIG. 5. The pressure coefficient $1/\rho(\partial\rho/\partial P)$ as a function of $T^{-1/2}$. The solid line is a guide to the eye.

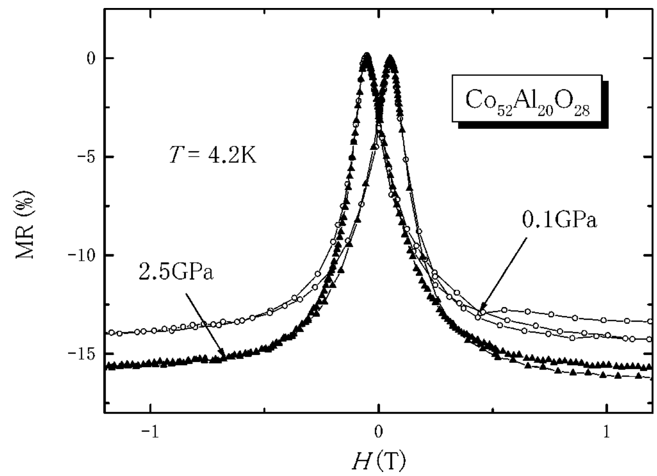

FIG. 6. MR curves measured at 4.2 K under 0.1 GPa and 2.5 GPa. The vertical axis indicates the values of $\Delta\rho/\rho_{max}$, $\Delta\rho=\rho(H)-\rho_{max}$.

threshold in Co-Al-O system ($\sim$20% oxygen). From the results of TEM observation and a suitable model calculation, $\langle s \rangle$ of 43 at. % Co was estimated to be about 50% larger than that of 52 at. % Co.[21] Considering that the transition from tunneling to metallic conduction occurs near the quantum resistance ($\sim$26 kΩ),[26] the critical resistivity in Co-Al-O is estimated to be about 10^{-2} Ω cm, which was confirmed experimentally.[20] In the present work, the resistivities of 43 at. % Co and 52 at. % Co are 0.5 Ω cm and 0.1 Ω cm at room temperature, respectively. This result indicates that 52 at. % Co is closer to the transition region than 43 at. % Co. Furthermore, it is observed that the 52 at. % Co shows a tendency of metallic conduction in the high-pressure range above 3 GPa.[27] On the basis of these facts, an instability in the electronic and structural properties is expected around the transition region. The large pressure effect on the electrical resistivity (see Fig.1) and the value of C of 52 at. % Co mentioned above probably reflects such instability.

Next we consider the results in Fig. 2. According to Eq. (1), the pressure coefficient of ρ is described as

$$\frac{1}{\rho}\frac{\partial\rho}{\partial P}\propto\frac{1}{\sqrt{T}}\frac{\partial(2\sqrt{C/k_B})}{\partial P}. \tag{2}$$

Since the pressure dependence of $2\sqrt{C/k_B}$ is found to be linear against pressure below 3.1 GPa as indicated in Fig. 4, $\partial(2\sqrt{C/k_B})/\partial P$ is constant. So the pressure coefficient is roughly proportional to $T^{-1/2}$, i.e., it increases with decreasing temperature. Figure 5 shows the pressure coefficient $1/\rho(\partial\rho/\partial P)$ as a function of $T^{-1/2}$ for 52 at. % Co. A linear relation is found in the temperature range below $T^{-1/2}\sim 0.2$ ($T=25$ K), but below 25 K, it shows a deviation from the linear dependence. In other words, the electrical conduction described by Eq. (1) is not applicable in this temperature range. A different conduction mechanism is needed to explain the conduction in this range. It is well established that the conduction of Co-Al-O granular materials in this temperature range is dominated by the higher-order (so-called cotunneling) effect in the tunnel conduction which will be mentioned in Sec. III C. In that section, we will also show that this effect becomes significant in low-temperature range below ca. 23 K, because the charging energy of granules (Coulomb blockade), which is as much as 23 K by theoretical calculation, plays an important role. Taking into account of these facts, it is suggested that the effect of higher-order tunneling is a possible origin for the deviation from $T^{-1/2}$ dependence in Fig. 5.

B. Tunneling magnetoresistance

Figure 6 shows the MR ratio, $\Delta\rho(H)/\rho_{max}$ at 4.2 K as a function of magnetic field H(T) at $P=0.1$ GPa and 2.5 GPa. $\Delta\rho(H)/\rho_{max}$ is defined as $\Delta\rho(H)/\rho_{max}=[\rho(H)-\rho_{max}]/\rho_{max}$, where ρ_{max} is the maximum resistivity around zero applied field and $\rho(H)$ is the resistivity at a field H. All MR curves show the maximum around 50 mT. As pressure increases, the MR ratio increases from 14% at 0.1 GPa to about 16% at 2.5 GPa. This result indicates that TMR is enhanced by applying pressure. Figure 7 shows the MR ratio

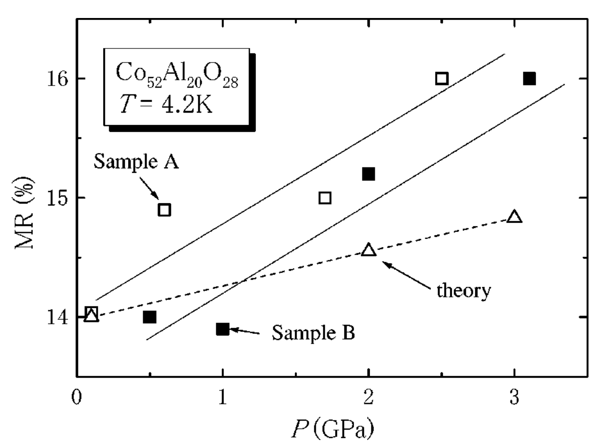

FIG. 7. Pressure dependence of the MR ratio for two samples A ($\square$) and B ($\blacksquare$). Triangles ($\triangle$) indicate MR ratios estimated from Eq. (5) for sample A at 2.0 GPa and 3.1 GPa. The dashed line shows the result of calculation.

at $T=4.2$ K as a function of pressure for samples, *A* and *B*. The values of TMR increase linearly with increasing pressure: the MR ratio increases by about 2% by applying 3.1 GPa. The pressure dependence of TMR of sample *A* is almost the same as that of *B*. In other words, there is no large difference between the samples as far as the pressure dependence of TMR is concerned. The TMR of 43 at. % Co was measured also at high pressure up to 2 GPa, in which the pressure dependence of TMR was found to be smaller than that of 52 at. % Co.

C. Higher-order tunneling

TMR in the granular systems at low temperatures has been reported by several authors.[28–33] It has been revealed that the Sheng's model and its extension to magnetic granular system incorporating the effect of spin-dependent tunneling is not able to explain the experimental results.[30] Recently, Takahashi and Maekawa[31] and Mitani *et al.*[29] presented a theory taking into account higher-order tunneling. In Sheng's model, tunneling is possible only between granules with the same size. On the other hand, the distribution in granular sizes[20] is well considered in the higher-order tunneling model: the large granules are separated by small ones due to their low number density. Since the effect of Coulomb blockade is significant at low temperature, higher-order processes become dominant in the conduction. The process is that a carrier is transferred from a charged large granules to a neighboring neutral large granules through the array of small granules, using the successive tunneling of electrons. According to this theory,[29] the temperature dependence of resistivity with the higher-order process is described as

$$\rho(T)=A'(1+m^2P^2)^{-(n^*+1)}(n^*/\tilde{\kappa}\langle s\rangle)^{-1/2} \times\frac{1}{f(n^*)}\exp[2\sqrt{2\tilde{\kappa}\langle s\rangle\langle E_c\rangle/k_BT}], \quad (3)$$

where $m=M/M_s$ is the magnetization normalized to the saturation magnetization M_s and P is spin polarization. $n^*=(\langle E_c\rangle/8\tilde{\kappa}\langle s\rangle k_BT)^{1/2}$, where $\tilde{\kappa}/\kappa\approx1+(1/4\kappa\langle s\rangle)\ln[(g/\pi)^2+(\langle E_c\rangle/2\pi k_BT)^2]$ and g is a constant. The function $f(n^*)$ represents a distribution of conduction paths, which is assumed to be $\propto 1/n^*$. $\langle s\rangle$ is a mean distance between granules with average size $\langle d\rangle$. Equation (3) shows temperature dependence similar to that in Eq. (1). Comparing Eq. (3) with Eq. (1), we obtain the coefficient C of $T^{-1/2}$ as

$$C=2\tilde{\kappa}\langle s\rangle\langle E_c\rangle. \quad (4)$$

The observed $\rho(T)$ can also be fitted to Eq. (3) with $m=0$. In Eq. (3), the unknown parameters are A', $\tilde{\kappa}\langle s\rangle$, and E_C. Choosing $A'=8.15\times10^{-2}$, $\tilde{\kappa}\langle s\rangle=4$, and $E_C/k_B=23$ K, $\rho(T)$ at ambient pressure is found to be well fitted at low temperatures. As mentioned above, the pressure effect on $\langle s\rangle$ is expected to be definitely larger than that on E_C, and $\tilde{\kappa}$ probably decreases with decreasing $\langle s\rangle$. Therefore, for simplicity, $\rho(T)$s at 2.1 and 3 GPa are fitted by adjusting only $\tilde{\kappa}\langle s\rangle$. The fitting curves shown by solid curves in Fig. 3 indicate that the observed pressure dependence of $\rho(T)$ is well described through the pressure effect on $\tilde{\kappa}\langle s\rangle$ in Eq. (3).

Using Eq. (3), the magnitude of TMR in granular systems is expressed as

$$\Delta\rho/\rho_{max}=1-(1+m^2P^2)^{-(n^*+1)} \quad (5)$$

$$\simeq(n^*+1)m^2P^2. \quad (6)$$

The temperature dependence of TMR in Co-Al-O granular films at ambient pressure has been well explained by this model.[29] TMR changes through the value of n^*: TMR increases when n^* becomes large and vice versa. n^* is related to the number of small granules between larger ones, which contribute to the higher-order tunneling. Equation (6) shows that the higher-order tunneling enhances TMR by the factor of (n^*+1). Taking the relation $n^*\propto(\langle E_C\rangle/\tilde{\kappa}\langle s\rangle)^{1/2}$ into account, n^* increases with increasing pressure because $\tilde{\kappa}\langle s\rangle$ decreases with pressure. This means that TMR in the higher-order tunneling regime is enhanced by applying pressure, which is in qualitative agreement with the present result in Fig. 7.

From Eq. (5), we have calculated the pressure-enhanced TMR, in which we used the same parameters as those in the fitting of $\rho(T)$. P was determined from the value of TMR at 0.1 GPa to be 0.315. The estimated values of TMR at high pressure on the basis of Eq. (6) are shown in Fig. 7 by a dashed line. The result shows an increase in TMR with increasing pressure, which is qualitatively in agreement with the present experimental results. The quantitative difference is probably due to the rough approximation in the simplified model. The present result indicates that the pressure-enhanced TMR is qualitatively explained by the higher-order tunneling theory.

IV. CONCLUSION

We have studied the effect of pressure on the electrical resistance and TMR of Co-Al-O granular thin film. The main results are summarized as follows.

(1) The steep increase in the $\rho(T)$ at low temperature is largely suppressed by applying pressure.

(2) The pressure coefficients of ρ show $T^{-1/2}$ dependence down to 25 K but a deviation from this is observed below 25 K.

(3) The magnitude of TMR at 4.2 K is enhanced by applying pressure.

The present results are understood qualitatively on the basis of the recent theory taking into account the higher-order tunneling and a decrease of the distance $\langle s\rangle$ between granules by applying pressure.

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